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Reformulation of the Dynamical Theory of Coherent Wave Propagation by Randomly Distorted Crystals

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Abstract

The coherent terms in the spherical-wave approach of the statistical dynamical theory are reformulated using rigorous boundary conditions in contrast to the intuitive boundary conditions of the original formulation by Kato [*Acta Cryst.* (1980), A36, 763-769, 770-778]. These boundary conditions are explained physically by a general interference effect between the forward-diffracted wave and the incident undiffracted wave (using the optical theorem) and their consequences on the total (Bragg and forward-diffracted) incoherent intensity are also discussed.

1. Introduction

The statistical dynamical theory (SDT) of Bragg diffraction by randomly distorted crystals, first formulated by Kato (1980*a*, *b*), can be divided into two parts. Only the first part, consisting in the calculation of the so-called coherent waves diffracted into the Bragg and forward directions, will be considered explicitly in the present paper. Even in the case of a nonabsorbing crystal, these coherent waves lose intensity, which is transferred to the incoherent beams calculated in the second part of the theory. We shall consider the case of a nonabsorbing crystal with a centrosymmetrical structure, in Laue symmetrical conditions. Our purpose is to reveal the differences between our approach and the previous one by Kato (1980*a*, *b*) and by Al Haddad & Becker (1988), Becker & Al-Haddad (1989, 1990, 1992). This is particularly clear when the static Debye-Waller factor is equal to zero; in the previous approach there are then no coherent diffracted intensities at all but in our approach there is indeed some coherent intensity in the forward-diffracted beam.

The present paper deals with the point-source functions (PSF) that represent the coherent waves generated by a point source on the entrance surface of the crystal (this is known as a 'spherical wave'). A slightly different form of the differential equations satisfied by the coherent PSF is proposed and special attention is paid to the boundary conditions which must be in agreement with the usual dynamical theory of diffraction by perfect and by nonrandomly deformed crystals. We show that the transmitted wave is not affected by the random distortion of the crystal in a narrow region close to the direction of the undiffracted wave. This has a physically meaningful consequence: the transmitted intensity is reduced by interference between the undiffracted wave and the forward-diffracted wave, this reduction being compensated for in the total diffracted intensity (this is simply a statement of the 'optical theorem' described in most textbooks on quantum mechanics).

Our results for the coherent PSF are in agreement with the paper by Polyakov, Chukhovskii & Piskunov (1991) and it is proposed to use a similar approach for the calculation of incoherent beams. This is discussed here and will be the topic of forthcoming papers.

2. The PSF in diffraction by a distorted crystal

Let O be a point source on the entrance surface of the crystal; we shall use the nonorthogonal coordinate system (OS_o, OS_h) shown in Fig. 1. Let $G_h(s_o, s_h)$ and $G_{od}(s_o, s_h)$ be the amplitudes of the Bragg-diffracted and forward-diffracted waves respectively.

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The total wave in the forward direction, including the incident wave represented by the delta function $\delta(s_h)$, is

$$G_o(s_o, s_h) = \delta(s_h) + G_{od}(s_o, s_h). \tag{1}$$

$G_o(s_o, s_h)$ and $G_h(s_o, s_h)$ are solutions of the Takagi differential equations

$$\begin{aligned} \partial G_h(s_o, s_h)/\partial s_h &= i\chi\varphi(s_o, s_h)G_o(s_o, s_h), \\ \partial G_o(s_o, s_h)/\partial s_o &= i\chi\varphi^*(s_o, s_h)G_h(s_o, s_h). \end{aligned} \tag{2}$$

Here, χ is the reciprocal of the so-called extinction length of the considered reflection and $\varphi(s_o, s_h)$ is the lattice phase factor

$$\varphi(s_o, s_h) = \exp[-i\mathbf{h}\cdot\mathbf{u}(s_o, s_h)],$$

which depends on the diffraction vector $\mathbf{h}$ and on the local displacement field $\mathbf{u}(s_o, s_h)$ of the crystal lattice. Equations (2) can be written in integral form,

$$\begin{aligned} G_h(s_o, s_h) &= i\chi \int_0^{s_h} \mathrm{d}\eta\, \varphi(s_o, \eta)G_o(s_o, \eta), \\ G_o(s_o, s_h) &= \delta(s_h) + i\chi \int_0^{s_o} \mathrm{d}\xi\, \varphi^*(\xi, s_h)G_h(\xi, s_h). \end{aligned} \tag{3}$$

Their solution can be represented by the following expansions, obtained by iteration:

$$\begin{aligned} G_h(s_o, s_h) &= \sum_{n=0}^{\infty} G_h^{(2n+1)}(s_o, s_h), \\ G_o(s_o, s_h) &= \delta(s_h) + \sum_{n=1}^{\infty} G_o^{(2n)}(s_o, s_h). \end{aligned} \tag{4}$$

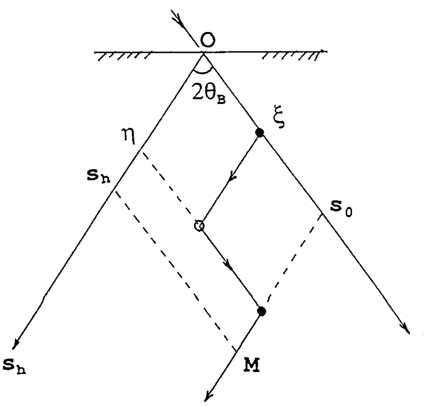

Fig. 1. Illustration of the spherical-wave geometry in the Laue case. θ_B is the Bragg angle. A 'zig-zag' path contributing to the $G_h^{(3)}(s_o, s_h)$ term of the Bragg-diffracted wave $G_h(s_o, s_h)$ is shown here. The symbols ● and ○ indicate the positions of scattering events from the forward o beam to the Bragg h beam and *vice versa*. The wave amplitudes $G_{o,h}(s_o, s_h)$ are zero outside the 'Borrmann fan' limited by the axes Os_o and Os_h. The total phase factor associated with this path is $\varphi(\xi, 0)\varphi^*(\xi, \eta)\varphi(s_o, \eta)$.

The first terms are

$$G_h^{(1)}(s_o, s_h) = i\chi \int_0^{s_h} \mathrm{d}\eta\, \varphi(s_o, \eta)\delta(\eta) = i\chi\varphi(s_o, 0),$$

$$G_o^{(2)}(s_o, s_h) = (i\chi)^2 \int_0^{s_o} \mathrm{d}\xi\, \varphi^*(\xi, s_h)\varphi(\xi, 0),$$

$$G_h^{(3)}(s_o, s_h) = (i\chi)^3 \int_0^{s_h} \mathrm{d}\eta \int_0^{s_o} \mathrm{d}\xi\, \varphi(s_o, \eta) \times \varphi^*(\xi, \eta)\varphi(\xi, 0).$$

The upper index (m) is the number of scattering events taking place between the point source and the observation point.

It should be noted that $G_h^{(1)}$ and $G_0^{(2)}$ are the only nonzero terms for $s_h = 0$:

$$\begin{aligned} G_h(s_o, 0) &= G_h^{(1)}(s_o, 0) = i\chi\varphi(s_o, 0) \\ G_{od}(s_o, 0) &= G_o^{(2)}(s_o, 0) = -\chi^2 s_o. \end{aligned} \tag{5}$$

This shows that $G_{od}(s_o, 0)$ does not depend on the crystal distortion.

In the case of a perfect crystal the successive terms are easily calculated and we thus recognize the well known solution in terms of Bessel functions:

$$\begin{aligned} G_h(s_o, s_h) &= \sum_0^{\infty} (i\chi)^{2n+1} s_o^n s_h^n / n!n! \\ &= i\chi J_0[2\chi(s_o s_h)^{1/2}], \\ G_{od}(s_o, s_h) &= \sum_1^{\infty} (i\chi)^{2n} s_o^n s_h^{n-1} / n!(n-1)! \\ &= \chi(s_o/s_h)^{1/2} J_1[2\chi(s_o s_h)^{1/2}]. \end{aligned} \tag{6}$$

3. The PSF in diffraction by a randomly distorted crystal

Following Kato (1980*a*), we now consider the lattice phase factor as a random function, from which a uniform static Debye–Waller factor E and a second-order correlation function $g(t)$ are defined such that

$$\langle\varphi(s_o, s_h)\rangle = E$$

$$\begin{aligned} \langle\varphi(s_o, s_h)\varphi^*(s_o \pm t, s_h)\rangle &= \langle\varphi(s_o, s_h)\varphi^*(s_o, s_h \pm t)\rangle \\ &= E^2 + (1 - E^2)g(t). \end{aligned} \tag{7}$$

A correlation length τ is defined as

$$\tau = \int_0^{\infty} \mathrm{d}t\, g(t)$$

We shall assume $\chi\tau \ll 1$. A convenient choice for $g(t)$ is

$$g(t) = \exp(-|t|/\tau). \tag{8}$$

The coherent wavefunctions are defined as the averaged quantities $\langle G_h(s_o, s_h)\rangle$ and $\langle G_o(s_o, s_h)\rangle$. The first terms of their expansions are

$$\langle G_h^{(1)}(s_o, s_h)\rangle = i\chi E$$
$$\langle G_{od}^{(2)}(s_o, s_h)\rangle = -\chi^2[E^2+(1-E^2)g(s_h)]s_o. \quad (9)$$

To obtain differential equations for $\langle G_h(s_o, s_h)\rangle$ and $\langle G_{od}(s_o, s_h)\rangle$, we consider the derivatives

$$\partial\langle G_h^{(2n+1)}(s_o, s_h)\rangle/\partial s_h = i\chi\langle\varphi(s_o, s_h)G_o^{(2n)}(s_o, s_h)\rangle = i\chi E\langle G_o^{(2n)}(s_o, s_h)\rangle - \chi^2\int_0^{s_o} d\xi\langle[\varphi(s_o, s_h)-E] \times \varphi^*(\xi, s_h)G_h^{(2n-1)}(\xi, s_h)\rangle.$$

We now introduce the decoupling approximation

$$\langle[\varphi(s_o, s_h)-E]\varphi^*(\xi, s_h)G_h^{(2n-1)}(\xi, s_h)\rangle = \langle[\varphi(s_o, s_h)-E]\varphi^*(\xi, s_h)\rangle\langle G_h^{(2n-1)}(\xi, s_h)\rangle.$$

Since we have

$$\langle[\varphi(s_o, s_h)-E]\varphi^*(\xi, s_h)\rangle = (1-E^2)g(s_o-\xi),$$

we obtain

$$\partial\langle G_h^{(2n+1)}\rangle/\partial s_h = i\chi E\langle G_o^{(2n)}\rangle - (1-E^2)\chi^2 \times \int_0^{s_o} d\xi\, g(s_o-\xi)\langle G_h^{(2n-1)}(\xi, s_h)\rangle$$

and, similarly,

$$\partial\langle G_o^{(2n)}\rangle/\partial s_o = i\chi E\langle G_h^{(2n-1)}\rangle - (1-E^2)\chi^2 \times \int_0^{s_h} d\eta\, g(s_h-\eta)\langle G_o^{(2n-2)}(s_o, \eta)\rangle$$

$$\partial\langle G_o^{(2)}\rangle/\partial s_o = i\chi E\langle G_h^{(1)}\rangle - (1-E^2)\chi^2 g(s_h).$$

In the remainder of this paper, we shall omit the symbol $\langle\ldots\rangle$. G_o and G_h thus denote the mean averaged (coherent) PSF.

We obtain, by recombining equations corresponding to different orders, the integro-differential equations

$$\partial G_{od}/\partial s_o = i\chi E G_h(s_o, s_h) - (1-E^2)\chi^2\int_0^{s_h} d\eta\, g(s_h-\eta)G_{od}(s_o, \eta) - (1-E^2)\chi^2 g(s_h), \quad (10)$$
$$\partial G_h/\partial s_h = i\chi E G_{od}(s_o, s_h) - (1-E^2)\chi^2\int_0^{s_o} d\xi\, g(s_o-\xi)G_h(\xi, s_h).$$

In the next section we shall use equations for G_o and G_h instead of G_{od} and G_h. We then obtain, instead of (10),

$$\partial G_o/\partial s_o = i\chi E G_h(s_o, s_h) + i\chi E\delta(s_h) - (1-E^2)\chi^2\int_0^{s_h} d\eta\, g(s_h-\eta)G_o(s_o, \eta)$$
$$\partial G_h/\partial s_h = i\chi E G_o(s_o, s_h) - (1-E^2)\chi^2\int_0^{s_o} d\xi\, g(s_o-\xi)G_h(\xi, s_h). \quad (11)$$

The last term of the first equation in (10) is not present in the corresponding equation of Kato (1980*a*). This is an important difference since we thus obtain, for $s_h = 0$,

$$\partial G_{od}(s_o, 0)/\partial s_o = -\chi^2 E^2 - (1-E^2)\chi^2 g(0) = -\chi^2,$$

which is in agreement with the boundary values (5).

For $s_o, s_h \gg \tau$, and since $\chi\tau \ll 1$, we get simple differential equations,

$$\partial G_{od}/\partial s_o = i\chi E G_h - \mu G_{od},$$
$$\partial G_h/\partial s_h = i\chi E G_{od} - \mu G_h, \quad (12)$$

with $\mu = (1-E^2)\chi^2\tau$, from which Kato proposed his solution for the coherent PSF:

$$G_{od} = -\chi E(s_o/s_h)^{1/2}J_1[2\chi E(s_o s_h)^{1/2}] \times \exp[-\mu(s_o+s_h)], \quad (13)$$
$$G_h = i\chi E J_o[2\chi E(s_o s_h)^{1/2}]\exp[-\mu(s_o+s_h)].$$

It will be shown that these expressions, which are not in agreement with the exact boundary conditions, may nevertheless be correct in the central part of the Borrmann fan.

4. Two-dimensional Laplace transformation of the coherent wave fields

We may apply a two-dimensional Laplace transformation to (11), corresponding to the coherent fields including the undiffracted part $\delta(s_h)$ in $G_o(s_o, s_h)$, to obtain

$$[p_o+(1-E^2)\chi^2\bar{g}(p_h)]G_o(p_o, p_h) - i\chi E G_h(p_o, p_h) = 1$$
$$-i\chi E G_o(p_o, p_h) + [p_h+(1-E^2)\chi^2\bar{g}(p_o)] \times G_h(p_o, p_h) = 0 \quad (14)$$

where

$$\bar{G}_{o,h}(p_o, p_h) = \iint_0^\infty ds_o\, ds_h \exp(-p_h s_h - p_o s_o) \times G_{o,h}(s_o, s_h)$$

are the two-dimensional transforms of G_o, G_h and

$\bar{g}(p_o)$, $\bar{g}(p_h)$ are one-dimensional transforms. From (8),

$$\bar{g}(p) = \int_0^\infty ds \exp(-ps) g(s)$$
$$= \int_0^\infty ds \exp(-ps) \exp(-s/\tau)$$
$$= \tau/(1+p\tau).$$

The solution of (11) is then, with $\mu = (1-E^2)\chi^2\tau$,

$$\begin{aligned} G_o(p_o, p_h) &= [p_h + \mu/(1+\tau p_o)] \\ &\quad \times \{\chi^2 E^2 + [p_o + \mu/(1+\tau p_h)] \\ &\quad \times [p_h + \mu/(1+\tau p_o)]\}^{-1} \\ G_h(p_o, p_h) &= i\chi E\{\chi^2 E^2 + [p_o + \mu/(1+\tau p_h)] \\ &\quad \times [p_h + \mu/(1+\tau p_o)]\}^{-1}. \end{aligned} \tag{15}$$

It does not appear possible to perform the two-dimensional inverse transformation analytically, so we will introduce approximations, except in the case $E=0$ for which an exact result is obtained. We have

$$G_o = [p_o + \chi^2/(p_h + 1/\tau)]^{-1}$$

so

$$\begin{aligned} G_o &\simeq \delta(s_h) - \chi(s_o/s_h)^{1/2} J_1[2\chi(s_o s_h)^{1/2}] \\ &\quad \times \exp(-s_h/\tau) \end{aligned} \tag{16}$$

and, if $E=0$,

$$G_h(s_o, s_h) = 0.$$

5. Approximations in different regions if $E \neq 0$

5.1. *The region $s_o, s_h \gg \tau$*

In this case, it is possible to assume in (14) that $\tau p_h \ll 1$ and $\tau p_o \ll 1$ so

$$p_o + \mu/(1+\tau p_h) \simeq p_o + \mu;$$
$$p_h + \mu/(1+\tau p_o) \simeq p_h + \mu,$$

hence

$$\begin{aligned} G_o(p_o, p_h) &= [p_o + \mu + \chi^2 E^2/(p_h+\mu)]^{-1}, \\ G_h(p_o, p_h) &= i\chi E[\chi^2 E^2 + (p_o+\mu)(p_h+\mu)]^{-1}. \end{aligned} \tag{17}$$

The inverse Laplace transformation of these expressions can be performed easily and leads to the simple expressions (13). In fact, Kato (1980*a*) did state that $s_o \gg \tau$ and $s_h \gg \tau$ are necessary conditions for these expressions to be valid.

According to Polyakov *et al* (1991), it is possible to use a less restrictive approximation in (15):

$$p_o + \mu/(1+\tau p_h) \simeq p_o + \mu - \mu\tau p_h,$$
$$p_h + \mu/(1+\tau p_o) \simeq p_h + \mu - \mu\tau p_o.$$

The inverse transformation is then more complicated but gives, without further approximation, the modified expressions

$$\begin{aligned} G_h &= [1-(\mu\tau)^2]^{-1} \exp[-(\mu+\mu^2\tau)(s_o+s_h)] \\ &\quad \times i\chi E J_o[2\chi E(s_o+\mu\tau s_h)^{1/2}(s_h+\mu\tau s_o)^{1/2}], \\ G_o &= -\chi E[1-(\mu\tau)^2]^{-1} \exp[-(\mu+\mu^2\tau)(s_o+s_h)] \\ &\quad \times [(s_o+\mu\tau s_h)/(s_h+\mu\tau s_o)]^{1/2} \\ &\quad \times J_1[2\chi E(s_o+\mu\tau s_h)^{1/2}(s_h+\mu\tau s_o)^{1/2}], \end{aligned} \tag{18}$$

which can be shown to be equivalent to (13) if

$$\chi\mu E\tau(s_h^{3/2}/s_o^{1/2} + s_o^{3/2}/s_h^{1/2}) \ll 1. \tag{19}$$

5.2. *The region $s_h \ll \tau$, $s_o \gg \tau$*

For this region we can simplify (15) by using the approximations

$$p_o\tau + 1 \simeq 1 \quad (p_o\tau \ll 1),$$
$$p_h\tau + 1 \simeq p_h\tau \quad (p_h\tau \gg 1),$$

so

$$G_o(p_o, p_h) = (p_h+\mu)[\chi^2 E^2 + (p_o + \mu/\tau p_h)(p_h+\mu)]^{-1}$$
$$G_h(p_o, p_h) = i\chi E[\chi^2 E^2 + (p_o + \mu/\tau p_h)(p_h+\mu)]^{-1}.$$

We make use of the fact that

$$\chi^2 E^2 + \mu/\tau = \chi^2 E^2 + (1-E^2)\chi^2 = \chi^2$$

and must make the approximation

$$p_h + \mu \simeq p_h$$

(which is valid since $\mu \ll 1/\tau$) then

$$\begin{aligned} G_o(p_o, p_h) &= p_h/(\chi^2 + p_o p_h), \\ G_h(p_o, p_h) &= i\chi E/(\chi^2 + p_o p_h). \end{aligned} \tag{20}$$

This is the solution of the perfect-crystal case

$$\begin{aligned} G_o(s_o, s_h) &= \delta(s_h) - \chi(s_o/s_h)^{1/2} J_1[2\chi(s_o s_h)^{1/2}], \\ G_h(s_o, s_h) &= i\chi E J_o[2\chi(s_o s_h)^{1/2}], \end{aligned} \tag{21}$$

except for the presence of the factor E in the expression for $G_h(s_o, s_h)$ (*cf.* Polyakov *et al.*, 1991).

5.3. *An approximation valid for any value of s_h and requiring only $s_o \gg \tau$*

In this case, we do not impose a restriction on p_h, but we can use

$$p_h + \mu/(1+\tau p_o) \simeq p_h + \mu \quad (\tau p_o \ll 1)$$

so that

$$\begin{aligned} G_o(p_o, p_h) &= [p_o + \mu/(1+\tau p_h) + \chi^2 E^2/(p_o+\mu)]^{-1}, \\ G_h(p_o, p_h) &= [i\chi E/(p_h+\mu)] \\ &\quad \times [p_o + \mu/(1+\tau p_h) + \chi^2 E^2/(p_o+\mu)]^{-1}. \end{aligned} \tag{22}$$

The inverse transformation involving p_o gives

$$\begin{aligned} G_o(s_o, p_h) &= \exp[-s_o\chi^2E^2/(p_h+\mu)] \\ &\quad\times\exp[-s_o\mu/(1+\tau p_h)], \\ G_h(s_o, p_h) &= i\chi E/(p_h+\mu) \\ &\quad\times\exp[-s_o\chi^2E^2/(p_h+\mu)] \\ &\quad\times\exp[-s_o\mu/(1+\tau p_h)]. \end{aligned} \tag{22'}$$

The inverse transformation involving p_h leads to convolution integrals

$$\begin{aligned} G_o(s_o, s_h) &= K(s_h) - \chi E s_o \int_0^{s_h} \mathrm{d}\eta(\exp[-\mu(s_h-\eta)] \\ &\quad\times J_1\{2\chi E[s_o(s_h-\eta)]^{1/2}\} \\ &\quad\times[s_o(s_h-\eta)]^{-1/2})K(\eta), \qquad (23) \\ G_h(s_o, s_h) &= i\chi E \int_0^{s_h} \mathrm{d}\eta \exp[-\mu(s_h-\eta)] \\ &\quad\times J_o\{2\chi E[s_o(s_h-\eta)]^{1/2}\}K(\eta), \end{aligned}$$

where $K(\eta)$ is the inverse Laplace transform of $\exp[-\mu s_o/(1+\tau p_h)]$,

$$\begin{aligned} K(\eta) &= \delta(\eta) - \exp(-\eta/\tau)(\mu s_o/\tau\eta)^{1/2} \\ &\quad\times J_1\{2[(\mu/\tau)s_o\eta]^{1/2}\} \end{aligned} \tag{24}$$

In fact, the present approximation breaks a symmetry relation resulting from the second equation of (15),

$$G_h(s_o, s_h) = G_h(s_h, s_o) \tag{25}$$

We propose to use (23) to calculate $G_h(s_o, s_h)$ if $s_o \gg s_h$ and the result of the calculation will also be taken as the value of G_h at the symmetrical point obtained by interchanging s_o and s_h.

The present approximation is in agreement with the rigorous formula (16) in the case $E=0$.

5.4. *A validity criterion for Kato's expressions*

Let us suppose first that $s_o > s_h > \tau$. The form of the function $K(\eta)$ is thus that the effective range of integration in (23) is aproximately $(0, \tau)$. The function $f(s_h-\eta)$ in (23), for G_0 or G_h, can be considered to have the constant value $f(s_h)$ when η varies from 0 to τ if the following conditions are satisfied:

$$\begin{aligned} &\mu\tau \ll 1; \\ &\tau\chi E(s_o/s_h)^{1/2} \ll 1. \end{aligned} \tag{26}$$

The result of (23) is then the product of $f(s_h)$ by the integral of $K(\eta)$. Since the upper limit of integration can be extended to ∞, the value of this integral is simply obtained by letting $p_h=0$ in the Laplace transform $\exp[-\mu s_o/(1+\tau p_h)]$ of $K(\eta)$. Then (23) is clearly reduced to the simple expressions (13) of Kato [since $s_h > \tau$ is assumed, the function $\delta(s_h)$ in the result for G_o can be omitted].

From (26), for the case $s_o \geq s_h$, and from the discussion of the preceding section, for the case $s_o < s_h$, we can use the criterion $\mu\tau \ll 1$, so can assume

$$\begin{aligned} &(1-E^2)\tau^2\chi^2 \ll 1 \\ &\tau\chi E[(s_o/s_h)^{1/2} + (s_h/s_o)^{1/2}] \ll 1, \end{aligned}$$

which are most easily satisfied at the centre of the Borrmann fan ($s_o \simeq s_h$).

6. Integrated intensities and interference effects along the $S_h=0$ direction

Besides the coherent waves considered in the preceding sections, there are also incoherent beams which, following Kato (1980*a*), can be separated into 'mixed incoherent' components, built from intensity first diffracted from the coherent *o* wave and *h* wave to the *h* and *o* incoherent beams, respectively, and 'pure incoherent' components built from intensity first diffracted directly from the incident wave to the incoherent *h* beam. The integrated intensities considered in the present section are obtained by integrating the intensity distributions (functions of the coordinates s_o and s_h) over the exit surface of the crystal and are therefore the sums of coherent, mixed incoherent and pure incoherent terms:

$$\begin{aligned} I_{od}^{\text{tot}} &= I_{od}^{\text{coh}} + I_o^{\text{mi}} + I_o^{\text{pi}} \\ I_h^{\text{tot}} &= I_h^{\text{coh}} + I_h^{\text{mi}} + I_h^{\text{pi}}. \end{aligned} \tag{27}$$

The previously existing calculations by Kato (1980*b*), Al Haddad & Becker (1988), Guigay (1989) and Becker & Al Haddad (1992) are such that

$$\begin{aligned} I_{od}^{\text{coh}} + I_h^{\text{coh}} &= E^2QT\exp(-2\mu T) \\ I_o^{\text{mi}} + I_h^{\text{mi}} &= (E^2Q/2\mu)[1-\exp(-2\mu T)] \\ &\quad - E^2QT\exp(-2\mu T) \\ I_o^{\text{pi}} + I_h^{\text{pi}} &= (1-E^2)(Q/2\mu)[1-\exp(-2\mu T)]. \end{aligned} \tag{28}$$

Consequently,

$$I_{od}^{\text{tot}} + I_h^{\text{tot}} = (Q/2\mu)[1-\exp(-2\mu T)]. \tag{29}$$

Here T is the crystal thickness along the OS_o direction and Q is the usual quantity $Q=(\lambda\chi^2/\sin 2\theta)$ such that the kinematical value of the diffracted intensity is equal to QT, λ being the wavelength of the incident radiation. μ is, as before, equal to $(1-E^2)\chi^2\tau$.

In the following we shall write the total incoherent intensities in the *o* beam and in the *h* beam as

$$\begin{aligned} I_o^{\text{inc}} &= I_o^{\text{mi}} + I_o^{\text{pi}}, \\ I_h^{\text{inc}} &= I_h^{\text{mi}} + I_h^{\text{pi}}. \end{aligned}$$

If the crystal is very thick ($\mu T \gg 1$), the coherent intensity disappears. The total intensity (29) then becomes entirely incoherent and saturates to the finite value $Q/2\mu$.

To show that (29), which actually results from an intuitive choice of the boundary conditions in SDT, is not satisfactory, we shall now use a rigorous approach based on a general rule known as the 'optical theorem' in wave scattering.

As shown in Fig. 2, an optical path of scattering order $2n$ contributing to the forward beam at a point $M(s_o, s_h)$ such that $s_h \ll \tau$ is made of n pairs of scattering points close to each other (separation $\ll \tau$). This results in phase cancellation in each pair and consequently along the whole path. This explains physically (Guigay, 1990) that the forward-diffracted beam is coherent and is the same as in the case of a perfect crystal in the region $s_h \ll \tau$, in agreement with (22).

Consequently, the incident undiffracted wave $\delta(s_h)$ and the forward-diffracted wave $G_{od}(s_o, s_h)$ interfere along $s_h = 0$. We must take this interference into account when we calculate the total intensity leaving the crystal. We easily obtain the interference term by using [*cf.* (21)]

$$G_{od}(s_o, s_h) = -\chi^2 s_o \theta(s_h) \quad \text{for } \chi^2 s_o s_h \ll 1 \qquad (30)$$

and considering the delta function $\delta(s_h)$ as the limit for $\varepsilon \to 0$ of the rectangle function that is equal to $1/\varepsilon$ for $-\varepsilon/2 < s_h < \varepsilon/2$ and zero elsewhere. The result is then $-\chi^2 s_o$ and is rewritten as $-QT$ since $s_o = T$ for $s_h = 0$ and we have to introduce the factor $\lambda/\sin 2\theta$ for calculation of the integrated intensity. The total diffracted intensity considered in (29) must have exactly the opposite value, $+QT$, to ensure that the total intensity leaving the crystal (diffracted and undiffracted) is exactly equal to the incident intensity.

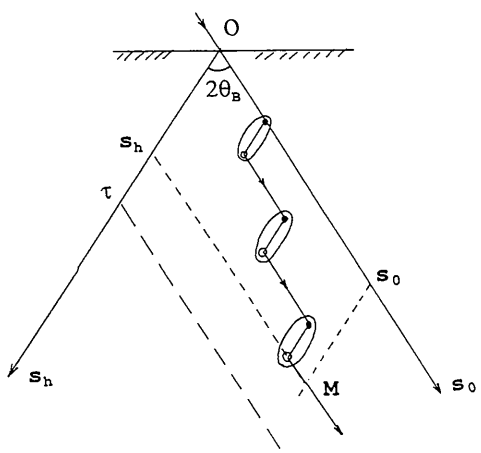

Fig. 2. A 'zig-zag' path contributing to the forward-diffreacted wave $G_{od}(s_o, s_h)$ with $s_h \lesssim \tau$. The successive pairs (● ○) of scattering points are such that the distance between the points of each pair is smaller than τ.

We thus get the simple relation

$$I_{od}^{\text{tot}} + I_h^{\text{tot}} = QT \qquad (31)$$

involving only the kinematical value of the integrated intensity, instead of (29).

7. Direct consequences for the incoherent intensities

7.1. *The case* $E = 0$

In this case, for which there is no I_h^{coh}, we have

$$I_o^{\text{inc}} + I_h^{\text{inc}} = QT - I_{od}^{\text{coh}}. \qquad (32)$$

I_{od}^{coh} can be calculated from (16) as the integral

$$I_{od}^{\text{coh}} = QT \int_{-1}^{1} \mathrm{d}\gamma \exp[-(T/\tau)(1-\gamma)] \times [(1+\gamma)/(1-\gamma)] J_1^2[\chi T(1-\gamma^2)^{1/2}], \qquad (33)$$

where γ is a dimensionless coordinate on the section of the Borrmann fan at the exit surface of the crystal and is such that

$$2s_h = T(1-\gamma),$$
$$2s_o = T(1+\gamma).$$

Since we suppose $T \gg \tau$ and because of the exponential term, only a small range of γ close to 1 is significant in (33). We substitute $\nu = 1-\gamma$, introduce simplifications of the integrand for $\nu \ll 1$ and take the integration range of ν from 0 to ∞ to obtain

$$I_{od}^{\text{coh}} = 2QT \int_0^{\infty} (\mathrm{d}\nu/\nu) \exp[-(T/\tau)\nu] J_1^2[\chi T(2\nu)^{1/2}].$$

This integral can be calculated exactly in terms of Bessel functions of purely imaginary arguments

$$I_{od}^{\text{coh}} = QT\{1 - \exp(-\chi^2\tau T)[I_o(\chi^2\tau T) + I_1(\chi^2\tau T)]\}. \qquad (34)$$

Then (32) can be written as

$$I_o^{\text{inc}} + I_h^{\text{inc}} = QT \exp(-\chi^2\tau T)[I_o(\chi^2\tau T) + I_1(\chi^2\tau T)], \qquad (35)$$

which differs from the result of Kato (1980*b*) and of Becker & Al Haddad (1990) in the case of $E = 0$,

$$I_o^{\text{inc}} + I_h^{\text{inc}} = QT[1 - \exp(-2\chi^2\tau T)]/2\chi^2\tau T.$$

In particular, for $\chi^2\tau T \gg 1$, using the asymptotic form of the functions $I_o(\varkappa)$ and $I_1(\varkappa)$ for large values $\varkappa \gg 1$,

$$I_o(\varkappa), I_1(\varkappa) \sim \exp(\varkappa)/(2\pi\varkappa)^{1/2},$$

we get

$$I_o^{\text{inc}} + I_h^{\text{inc}} \simeq \begin{cases} Q/2\chi^2\tau & \text{(previous theory)} \\ (Q/\chi)(T/2\pi\tau)^{1/2} & \text{(present theory).} \end{cases} \qquad (36)$$

We thus find that the total incoherent intensity increases as $T^{1/2}$ as a function of the crystal thickness instead of reaching a constant value.

7.2. *The general case*, $E \neq 0$

In this more complicated case, we shall only obtain the behaviour of $I_o^{\rm inc} + I_h^{\rm inc}$ for very thick crystals such that $\chi^2\tau T > 1$. Generally, we have

$$I_o^{\rm inc} + I_h^{\rm inc} = QT - I_{od}^{\rm coh} - I_h^{\rm coh}. \qquad (37)$$

We can calculate, using (21), the part of $I_{od}^{\rm coh}$ corresponding to the region $s_h < \tau$. We obtain an integral that can be calculated exactly,

$$Q\int_0^\tau {\rm d}\eta (T/\eta) J_1^2[2\chi(T\eta)^{1/2}]$$
$$= QT\{1 - J_0^2[2\chi(T\tau)^{1/2}] - J_1^2[2\chi(T\tau)^{1/2}]\}. \qquad (38)$$

For $\chi^2\tau T > 1$, this is close to the value QT of the total diffracted intensity (31). The coherent intensity is thus mainly concentrated in the region $s_h \ll \tau$. We can then consider (38) as the value of the total coherent intensity in (37). We get

$$I_o^{\rm inc} + I_h^{\rm inc} \simeq QT\{J_0^2[2\chi(T\tau)^{1/2}] + J_1^2[2\chi(T\tau)^{1/2}]\}$$
$$\simeq (Q/\pi\chi)(T/\tau)^{1/2} \qquad (39)$$

We thus obtain in the general case $E \neq 0$ for thick crystals a result similar to (36) in the $E = 0$ case.

8. Concluding remarks

Kato (1991) has recently presented a new discussion of the foundations of the statistical diffraction theory using wave equations that are more general than the Takagi–Taupin equations. The present paper is strictly related to the formulation of Kato (1980*a*, *b*), including the modifications introduced by Al Haddad & Becker (1988), by Becker & Al Haddad (1989, 1990) and by Guigay (1989). In this formulation the coherent waves have the simple form given in (13), which is expected to be a good approximation in the region around the middle of the Borrmann fan. The more general and yet simple expressions (18) have been obtained by Polyakov *et al.* (1991).

Starting from the two-dimensional Laplace transform of the coherent waves, we have obtained more rigorous expressions that are not so simple but are valid from the middle to the edges of the Borrmann fan. We hope that the expansion in terms of Bessel functions and Laguerre polynomials given in the Appendix may be useful for numerical calculations. Indeed, it would be interesting to investigate numerically the integrated intensities of the coherent beams and also their angular distribution; this problem has been considered recently by Bushuev (1989) in the case of a one-dimensional random crystal deformation.

In the present paper the boundary conditions have been considered rigorously and explained physically. In the boundary region $s_h < \tau$, the forward-diffracted wave is the same as for a perfect crystal, independently of the value of E. Consequently, the physically meaningful interference effect related to the optical theorem is of general validity. From this, we are able to show that the total diffracted incoherent intensity should increase as $(T/\tau)^{1/2}$ for large values of the thickness T such that $\chi^2\tau T \gg 1$, instead of reaching a finite limit as predicted by the previous formulation of the dynamical statistical theory. The distribution of this total incoherent intensity between the Bragg and the forward-diffracted beams will be considered in forthcoming papers; for this purpose, we intend to use a formalism based on the Bethe–Saltpeter equations, as suggested by Holy & Gabrielyan (1987) and by Polyakov *et al.* (1991).

APPENDIX

In our calculations, we have often used the following inverse Laplace transforms (m is a positive integer):

$$\exp(-a/p) \to \delta(t) - (a/t)^{1/2} J_1[2(at)^{1/2}]$$
$$p^{-m}\exp(-a/p) \to (t/a)^{(m-1)/2} J_{m-1}[2(at)^{1/2}]$$
$$p^{m}\exp(-a/p) \to \delta^{(m)}(t) + \ldots + [(-a)^m/m!]\delta(t) + (a/t)^{(m+1)/2} J_{-m-1}[2(at)^{1/2}].$$

These results are easily obtained from the series expansions of the exponential and Bessel functions. For $t > 0$, the delta function $\delta(t)$ and its derivatives may be omitted.

The integral expressions (23) can be represented by series expansions in terms of Laguerre polynomials and Bessel functions:

$$\begin{aligned} G_o(s_o, s_h) &= -\chi E \exp[\mu s_o/(1-\mu\tau) - \mu s_h] \\ &\quad \times (s_o/s_h)^{1/2} (J_1[2\chi E(s_o s_h)^{1/2}] \\ &\quad + \sum_{n=1}^{\infty} [\tau\chi E/(1-\mu\tau)]^n (s_o/s_h)^{n/2} \\ &\quad \times J_{n+1}[2\chi E(s_o s_h)^{1/2}] \\ &\quad \times \{L_n[\mu s_o/(1-\mu\tau)] \\ &\quad - L_{n-1}[\mu s_o/(1-\mu\tau)]\}) \\ G_h(s_o, s_h) &= i\chi E \exp[-\mu s_o/(1-\mu\tau) - \mu s_h] \\ &\quad \times (J_o[2\chi E(s_o s_h)^{1/2}] \\ &\quad + \sum_{n=1}^{\infty} [\tau\chi E/(1-\mu\tau)]^n (s_o/s_h)^{n/2} \\ &\quad \times J_n[2\chi E(s_o s_h)^{1/2}] \\ &\quad \times \{L_n[\mu s_o/(1-\mu\tau)] \\ &\quad - L_{n-1}[\mu s_o/(1-\mu\tau)]\}). \end{aligned} \qquad (40)$$

These expressions, which may be useful for numerical calculations, are obtained by using the generating function of the Laguerre polynomials $L_n(x)$:

$$\exp[-x/(1-t)] = (1-t)\exp(-x)\sum_{n=0}^{\infty} t^n L_n(x).$$

Indeed, in (22′), using

$$x_o = \mu s_o/(1-\mu\tau), \quad \tau' = \tau/(1-\mu\tau), \quad q = p_h + \mu,$$

we can write

$$\begin{aligned}\exp[-s_o\mu/(1+\tau p_h)] &= \exp[-x_o/(1+\tau' q)] \\ &= \exp(-x_o)(1+\tau' q)\sum_{n=0}^{\infty}(-\tau' q)^n L_n(x_o) \\ &= \exp(x_o)\left\{1+\sum_{n=0}^{\infty}(-\tau' q)^n[L_n(x_o)-L_{n-1}(x_o)]\right\}.\end{aligned}$$

The formulae (40) are then obtained by using the inverse Laplace transform of $q^n \exp(-s_o\chi^2E^2/q)$.

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The Observation of Phonons in Barium Fluoride by Pulsed Neutron Diffraction

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Abstract

Measurements of thermal diffuse scattering from a single crystal of barium fluoride made on the neutron time-of-flight Laue single-crystal diffractometer SXD are presented. These measurements are shown to confirm present theories on the nature of the processes producing this scattering effect and their striking variation with scattering geometry.

1. Introduction

In earlier papers (Willis, 1986; Schofield & Willis, 1987), we have discussed the nature of thermal diffuse scattering (TDS) which occurs close to the Bragg reflections in time-of-flight neutron diffraction. Some of the theoretical predictions arising from these studies have received experimental support from observations on pyrolytic graphite (Willis, Carlile, Ward, David & Johnson, 1986) and on single crystals of barium fluoride and calcium fluoride (Carlile & Willis, 1989). These experiments were performed on the high-resolution powder diffractometer (HRPD) at the ISIS Pulsed Neutron Facility, using scattering angles around $2\theta = 174°$. We have now carried out similar experiments with the single-crystal diffractometer (SXD) at ISIS using reduced scattering angles around $2\theta = 90°$ and $2\theta = 125.5°$. Some striking new features occur in the TDS pattern at these lower scattering angles. We shall give an account of these new observations and show that they can also be explained satisfactorily by theory.

2. Time-of-flight study of TDS from barium fluoride

The single-crystal diffractometer SXD at ISIS is a time-of-flight Laue instrument ideally suited to surveying measurements in reciprocal space. The instrument has a short primary flight path ($L_1 = 8$ m, compared with the HRPD with $L_1 \simeq 100$ m). In spite of the poorer resolution of the instrument ($\sim 5\times10^{-3}$ in $\Delta Q/Q$ compared with 5×10^{-4} for HRPD), it is straightforward to observe the TDS features of interest in this work.

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